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(54) Title: ALKYLAMINOACETAMIDE LUBRICANT ADDITIVES

(57) Abstract: The invention relates to a composition comprising a base oil of lubricating viscosity and alkylamine, alkylenedi-, alkylenetri- or alkylenetetra-amine derivatives of N-alkyl-halo-acetamides. These compounds are useful as ashless, phosphorus-free and sulfur-free anti-wear and friction modifying additives for lubricating oils.



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Alkylaminoacetamide Lubricant Additives

The invention relates to the use of alkylaminoacetamides, that is alkylamine, alkylenedi-, alkylenetri- or alkylenetetra-amine derivatives of N-alkyl-halo-acetamides, as ashless, phosphorus-free and sulfur-free antiwear and friction modifying additives for lubricating oils.

- 5 *U.S. Patent Specification Nos. 5,282,872 and 5,071,445* disclose amide, amide/ammonium salt or ammonium salt compounds of an aminoalkylene polycarboxylic acid and a secondary fatty amine as additives in fuels.

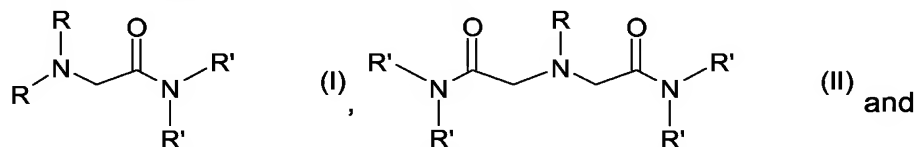
U.S. Patent Specification No. 5,376,155 teaches the reaction products of aminoalkylenecarboxylic acids with primary or secondary amines as paraffin dispersants in mineral oils.

- 10 It has now been found that certain N-alkylaminoacetamide or alkylenedi-, alkylenetri- or alkylenetetra-amine acetamide compounds are excellent antiwear and friction modifying additives for lubricating oils.

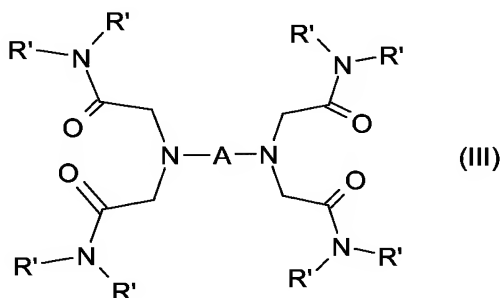
The invention relates to a composition, which comprises

- a) A base oil of lubricating viscosity, and
 15 b) An effective antiwear or friction modifying amount of at least one compound selected from the group consisting of

N-alkylaminoacetamide compounds (I), (II):



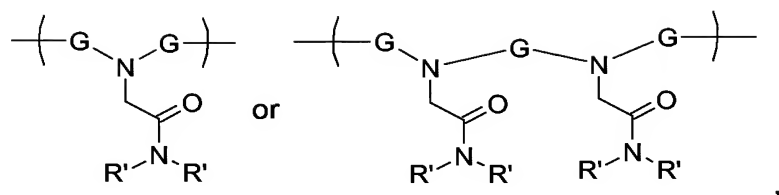
Alkylenedi-, alkylenetri- or alkylenetetra-amine acetamide compounds (III):



Wherein

A is alkylene of from 2 to 6 carbon atoms or is a group

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G, each independently, is alkylene of 2 to 6 carbon atoms,

R, each independently, is alkyl or alkenyl of 1 to 8 carbon atoms and

R', each independently, is hydrogen or alkyl or alkenyl or 1 to 24 carbon atoms,

5 provided that

each amide nitrogen atom is either mono- or disubstituted by alkyl or alkenyl; and that the alkyl or alkenyl groups have 8 to 24 carbon atoms, if the amide nitrogen is monosubstituted by alkyl or alkenyl; and

10 that the carbon atoms of the alkyl or alkenyl groups have 8 to 18 carbon atoms, if the amide nitrogen is disubstituted by alkyl or alkenyl. Claim 1

According to specific embodiment of the invention, the compounds (I), (II) or (III) are present from about 0.15% to about 10% by weight, based on the weight of the lubricant.

Component a)

15 A base oil of lubricating viscosity ("lubricant") can be used for the preparation of combustion engine oils. The total sulphur content in the low sulphur oil should not exceed the limit of more than 0.3 weight% with regard to the total weight of the composition.

Suitable combustion engine oils are based, for example, on mineral oils, natural oils, synthetic oils or mixtures thereof. These oils are known and familiar to the person skilled in the art and are described in standard reference books, such as in *Chemistry and Technology of*
 20 *Lubricants*; Mortier, R.M. and Orszulik, S.T. (Editors); 1992 Blackie and Son Ltd. for GB, VCH-Publishers N.Y. for U.S., ISBN 0-216-92921-0, pages 208 et seq. and 269 et seq.; in *Kirk-Othmer Encyclopedia of Chemical Technology, Fourth Edition 1969*, J. Wiley & Sons, New York, Vol. 13, page 533 et seq. (Hydraulic Fluids); *Performance Testing of Hydraulic Fluids*; R. Tourret and E.P. Wright, Hyden & Son Ltd. GB, on behalf of The Institute of Pe-
 25 *troleum London*, ISBN 0 85501 317 6; *Ullmann's Encyclopedia of Ind. Chem., Fifth Completely Revised Edition*, Verlag Chemie, DE-Weinheim, VCH-Publishers for U.S., Vol. A 15, page 423 et seq. (lubricants), Vol. A 13, page 165 et seq. (hydraulic fluids).

The base oil of lubricating viscosity is preferably a mineral oil derived lubricating base oil that contains 80% by mass or more of a saturated hydrocarbon component. Various methods for producing the mineral oil derived lubricating base oil are available. For example, the lubricating base oil may be a paraffin oil or a naphthenic oil obtainable by subjecting a lubricating oil fraction derived from an atmospheric or vacuum distillation of crude oil to refining processes such as deasphalting, solvent refining such as solvent extraction with furfural, hydrocracking, solvent or catalytic dewaxing, such as solvent or catalytic dewaxing, hydrotreating, such as hydrocracking or hydrofinishing, clay treatment, such as washing with acid treated or activated clay, or chemical refining such as washing with caustic soda or sulphuric acid and the like. Combinations of these methods are also available for producing the mineral oil derived lubricating base oil.

Component b)

According to a preferred embodiment, the invention relates to a composition, which comprises at least one compound selected from the group consisting of N-alkylaminoacetamide compounds (I) and (II), wherein the total number of carbon atoms of the alkyl or alkenyl groups are from 14 to 18 carbon atoms if the amide nitrogen is disubstituted by alkyl or alkenyl. Claim 2

According to another preferred embodiment, the invention relates to a composition, which comprises at least one compound selected from the group consisting of alkylenedi-, alkylenetri- and alkylenetetra-amine acetamide compounds (III), wherein A and G are selected from the group consisting of ethylene, propylene, hexamethylene and 2-methylpentylene. Claim 3

According to other preferred embodiments, the invention relates to compositions, wherein each R' is independently straight or branched chain alkyl or alkenyl of 7 to 9 carbon atoms claim 4; or

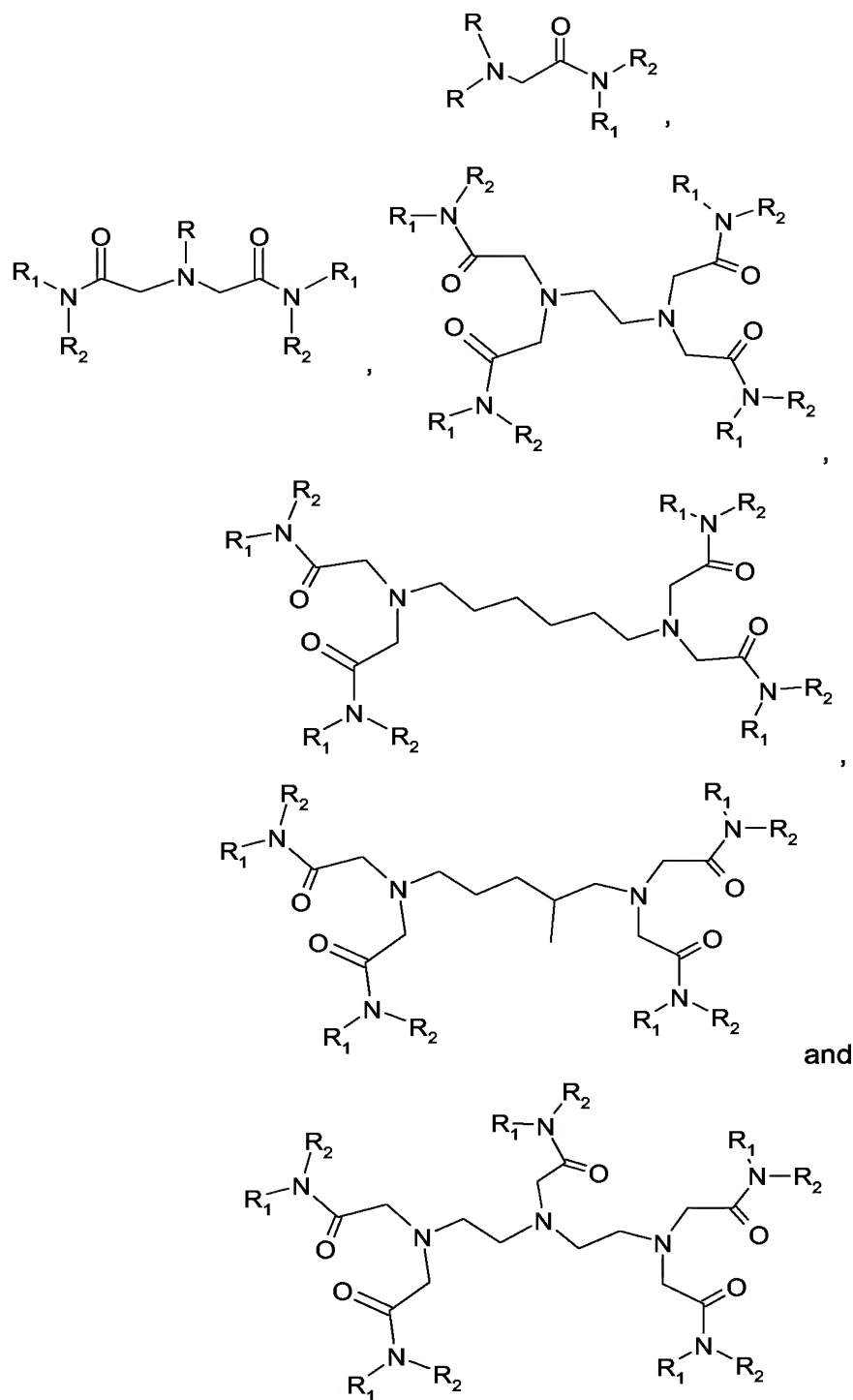
wherein one R' is hydrogen and the other one is straight or branched chain alkyl or alkenyl of 14 to 18 carbon atoms Claim 5; or

where each R' is identical and is straight or branched chain alkyl or alkenyl of 7 to 9 carbon atoms Claim 6.

For example, if an amide nitrogen is disubstituted by alkyl or alkenyl, the total carbon atoms of said alkyls or alkenyls combined are from 14 to 18 carbon atoms.

A particularly preferred embodiment relates to a composition, wherein the compounds (I), (II) and (III) are selected from the group consisting of

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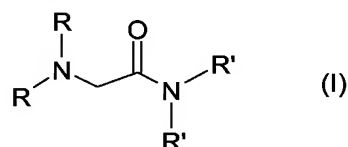


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wherein R is n-octyl and each of R₁ and R₂ are 2-ethylhexyl or n-octyl or one of R₁ and R₂ is hydrogen and the other is oleyl, n-octyl, t-octyl or dodecyl. Claim 7

- 5 -

A further embodiment relates to novel alkylaminoacetamide compounds (I)



Wherein

R, each independently, is alkyl or alkenyl of 1 to 8 carbon atoms and

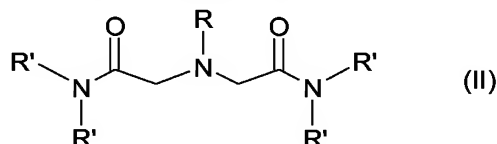
5 R', each independently, is hydrogen or alkyl or alkenyl or 1 to 24 carbon atoms,

provided that

each amide nitrogen atom is either mono- or disubstituted by alkyl or alkenyl; and that the alkyl or alkenyl groups have 8 to 24 carbon atoms, if the amide nitrogen is monosubstituted by alkyl or alkenyl; and

10 that the alkyl or alkenyl groups have 8 to 18 carbon atoms, if the amide nitrogen is disubstituted by alkyl or alkenyl. Claim 8

Another embodiment relates to novel N-alkylaminoacetamide compounds (II)



Wherein

15 R, each independently, is alkyl or alkenyl of 1 to 8 carbon atoms and

R', each independently, is hydrogen or alkyl or alkenyl or 1 to 24 carbon atoms,

provided that

each amide nitrogen atom is either mono- or disubstituted by alkyl or alkenyl; and that the alkyl or alkenyl groups have 8 to 24 carbon atoms, if the amide nitrogen is monosubstituted by alkyl or alkenyl; and

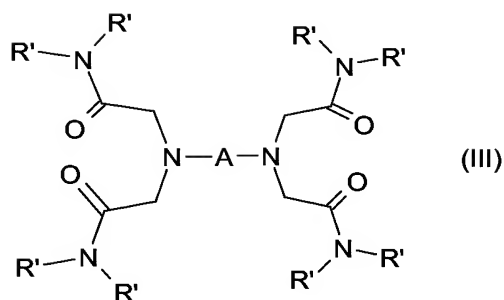
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that the alkyl or alkenyl groups have 8 to 18 carbon atoms, if the amide nitrogen is disubstituted by alkyl or alkenyl Claim 9 .

Another embodiment relates to novel alkylenedi-, alkylenetri- or alkylenetetra-amine acetamide compounds (III)

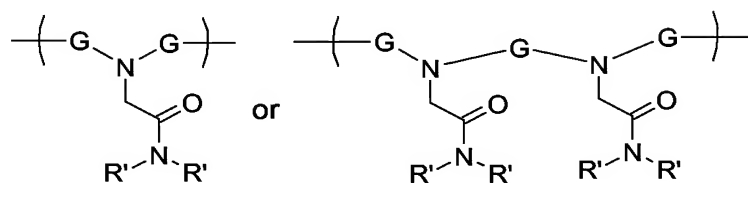
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- 6 -



wherein

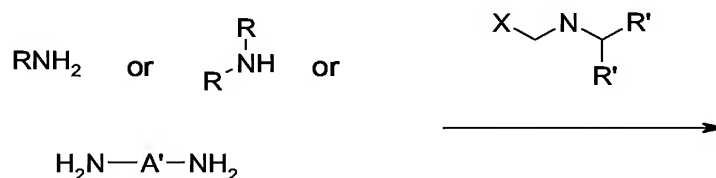
A is alkylene of from 2 to 6 carbon atoms or is a group



5 G, each independently, is alkylene of 2 to 6 carbon atoms,

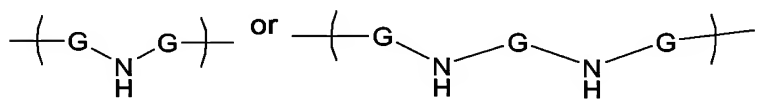
R', each independently, is alkyl or alkenyl of 1 to 24 carbon atoms, where for each amide group, the total number of carbon atoms of the alkyls or alkenyl groups are from 14 to 18 carbon atoms. Claim 10

10 The present N-alkylaminoacetamide compounds and alkylenedi-, alkylenetri- or alkylene-tetra-amine acetamide compounds are prepared for example by alkylation of primary or secondary amines with mono- or di-alkyl substituted halo-acetamides according to the following scheme:



where

15 A' is alkylene of from 2 to 6 carbon atoms or is a group



X is Cl, Br or I and R, R' and G are as previously defined.

Chloroacetamides are described in for example *U.S. Patent Specification. Nos. 2,746,901* and *4,801,618*. The compounds (I) and (II) may be prepared according to methods disclosed in these references. Haloacetamides are also described by *van Esch, et al., J. Org. Chem. 1995, 60, 1599-1610*, and by *Weaver, et al., J. Am. Chem. Soc. 1947, 69, 515-516*.

- 5 The compounds (I), (II) and (III) are prepared by reacting a haloacetamide with an appropriate amine or di-, tri- or tetra-amine in the ratio of about 1 molar equivalent of haloacetamide per reactive aminic hydrogen. For example, butylamine has two reactive aminic hydrogen atoms, dibutylamine has one reactive aminic hydrogen, and ethylenediamine has four reactive aminic hydrogen atoms. The reagents are mixed neat or in a suitable solvent, at a suitable
10 temperature and for a suitable time to complete the reaction. The reagents are mixed in the presence of an inorganic base, for example sodium carbonate.

Presumably, the compounds prepared in this way are not salts. They do not contain any ammonium salts. They do not contain any ammonium carboxylate salts, which would exist if prepared for example from the reaction of an alkylamine and an aminoalkylcarboxylic acid.

- 15 In the additives (I), (II) or (III), R is for example alkyl or alkenyl of 5 to 8 carbon atoms. A and G are, for example, ethylene, propylene, hexamethylene or 2-methylpentylene. For example, each amide is disubstituted by alkyl or alkenyl. For example, each R' is the same. For example, each R' is straight or branched chain alkyl or alkenyl of 7 to 9 carbon atoms. For example, one R' of each amide is hydrogen and the other is straight or branched chain alkyl or al-
20 kenyl of 14 to 18 carbon atoms.

For example, in the additives, each of the amide nitrogens are disubstituted by alkyl or alkenyl and the total carbon atoms of said alkyls or alkenyls for each amide combined are from 14 to 18 carbon atoms, that is to say, each R' is straight or branched chain alkyl or alkenyl of 7 to 9 carbon atoms.

- 25 For instance, in the additives, R is n-octyl and each R' is 2-ethylhexyl or n-octyl or one R' of each amide is hydrogen and the other is oleyl, n-octyl, t-octyl or dodecyl.

- Alkyl is straight or branched chain and is for example methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methyl-
30 heptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, icosyl or docosyl.

Alkenyl is also straight or branched and is an ethylenically unsaturated version of alkyl, for example allyl, oleyl, docosenyl, and the like.

Alkylene is straight or branched and is for example ethylene, propylene, methylethylene, tetramethylene, pentamethylene or hexamethylene.

Suitable amines are for example octylamine, dioctylamine, butylamine, dibutylamine, ethylenediamine, diethylenetriamine, dipropylenetriamine, $\text{H}_2\text{N}-(\text{CH}_2)_3\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{NH}_2$, and the like.

The additive compounds (I), (II) and (III), that is the additives, are highly soluble in the lubricant, for example are soluble at 1%, 2% or for example at 5% on a weight/weight basis at room temperature.

The additives, when employed in lubricants (lubricating oils) for internal combustion engines, serve both to reduce wear of the engine's moving parts and to reduce friction. The additives provide an economic method to accomplish desired antiwear and reduced friction properties without the use of a metal such as Zn, or the elements S or P, enabling a significant reduction or the elimination of P, S, and Zn containing additives.

The lubricants of component a) are, for example, those employed in internal combustion engines. The lubricants have necessary lubricating viscosity and are for example mineral oils or are synthetic and mixtures thereof.

Greases or other solid lubricants are also lubricating oils according to this invention.

Synthetic hydrocarbon oils include long chain alkanes such as cetanes and olefin polymers such as trimer and tetramers of octane and decene. These synthetic oils can be mixed with 1) ester oils such as pentaerythritol esters of monocarboxylic acids having about 2 to 20 carbon atoms, 2) polyglycol ethers, 3) polyacetals and 4) siloxane fluids. Useful among the synthetic esters are those made from polycarboxylic acids and monohydric alcohols. For example, ester fluids made from pentaerythritol or mixtures thereof with di- and tripentaerythritol, and an aliphatic monocarboxylic acid containing from 1 to 20 carbon atoms, or mixtures of such acids. Other examples are ester fluids made from trimethylolpropane and an aliphatic monocarboxylic acid containing from 1 to 20 carbon atoms, or mixtures of such acids.

The lubricating oils are also for example crude oil, industrial lubrication oils, cutting oil, metal working fluids and greases.

The additives of this invention are advantageously present in the lubricant at a level of for example from about 0.15% to about 10.0% by weight of lubricant. For example, the additives are present from about 0.15% to about 7.0%, from about 0.25% to about 5.0%, from about 0.5% to about 3.0%, or from about 0.75% to about 2.0% by weight of the lubricant. For ex-

ample, the additives are present from about 0.5% to about 5.0%, from about 0.5% to about 7.0%, or from about 0.5% to about 10.0% by weight, based on the weight of the lubricant.

In the event that in lubricating compositions operated under extremely adverse conditions, such as lubricating compositions for marine diesel engines, that the additives of this invention
5 may be present in amounts of up to about 30.0% by weight, or more, of the total weight of the lubricating composition.

The lubricating oils in accordance with the invention may additionally include other additives, which are added in order to improve still further the basic properties of these formulations; such additives include antioxidants, metal passivators, rust inhibitors, corrosion inhibitors,
10 viscosity index improvers, extreme pressure agents, pour point depressants, solid lubricants, dispersants, detergents, antifoams, color stabilizers, further high-pressure additives, demulsifiers, antiwear additives and additives which reduce the coefficient of friction. Such additives are added in the customary amounts in each case in the range from in each case about 0.01% to 10.0% by weight, based on the lubricating oil.

15 The list below gives some representative examples of such additional additives:

Examples of antioxidants:

- 1) Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-iso-butylphenol, 2,6-di-cyclopentyl-4-methylphenol, 2-(α -methyl-cyclohexyl)-4,6-
20 dimethylphenol, 2,6-di-octadecyl-4-methylphenol, 2,4,6-tri-cyclo-hexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, linear or sidechain-branched nonylphenols, for example 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methyl-undec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol or mixtures thereof;
- 25 2) Alkylthiomethylphenols, for example 2,4-di-octylthiomethyl-6-tert-butylphenol, 2,4-di-octylthiomethyl-6-methylphenol, 2,4-di-octylthiomethyl-6-ethylphenol or 2,6-di-dodecylthiomethyl-4-nonylphenol;
- 3) Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butyl-hydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-
30 tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate or bis(3,5-di-tert-butyl-4-hydroxyphenyl) adipate;
- 4) Tocopherols, for example α -, β -, γ - or δ -tocopherol or mixtures thereof (vitamin E);

- 5) Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis-(6-tert-butyl-2-methylphenol), 4,4'-thiobis(3,6-di-sec-amylphenol) or 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl) disulfide;
- 5 6) Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis(4-methyl-6-(alpha-methylcyclohexyl)-phenol), 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis(6-
10 (alpha-methylbenzyl)-4-nonylphenol), 2,2'-methylene-bis(6-(alpha,alpha-dimethylbenzyl)-4-nonylphenol), 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobu-
15 tane, ethylene glycol bis(3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate), bis(3-tert-butyl-4-hydroxy-5-methylphenyl)dicyclo-pentadiene, bis(2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl)terephthalate, 1,1-bis(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptobutane or 1,1,5,5-tetra(5-tert-butyl-4-hydroxy-2-
20 methylphenyl)-pentane;
- 7) O- N- and S-Benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl 4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl 4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl) sul-
25 fide or isooctyl 3,5-di-tert-butyl-4-hydroxy-benzylmercaptoacetate;
- 8) Hydroxybenzylated malonates, for example-dioctadecyl 2,2-bis(3,5-di-tert-butyl-2-hydroxybenzyl)malonate, dioctadecyl 2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)-malonate, di-dodecyl mercaptoethyl-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-malonate or di(4-(1,1,3,3-tetramethylbutyl)phenyl)2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate;
- 30 9) Aromatic hydroxybenzyl compounds, for example 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene or 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol;
- 10) Triazine compounds, for example 2,4-bisooctylmercapto-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triaz-

- ine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl) isocyanurate, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexahydro-1,3,5-triazine or 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)-isocyanurate;
- 5
- 11) Benzylphosphonates, for example dimethyl 2,5-di-tert-butyl-4-hydroxybenzyl-phosphonate, diethyl 3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl 3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl 5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate or the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid;
- 10
- 12) Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide or octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate;
- 13) Esters of β -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid, β -(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid, β -(3,5-dicyclohexyl-4-hydroxyphenyl)-propionic acid, 3,5-di-tert-butyl-4-hydroxyphenylacetic acid or β -(5-tert-butyl-4-hydroxyphenyl)-3-thiabutyric acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxalamide, 3-thiaundecanol, 3-thiapentadecanol, trimethyl-hexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo(2.2.2)octane, glycerol or transesterification products based on natural triglycerides of, for example, coconut oil, rape seed oil, sunflower oil or colza oil;
- 15
- 20
- 14) Amides of β -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid, e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamine or N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazine;
- 25
- 15) Ascorbic acid (vitamin C);
- 16) Amine-type antioxidants, for example N,N'-diisopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methyl-pentyl)-p-phenylenediamine, N,N'-bis(1-methyl-heptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-di-(naphth-2-yl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phen-
- 30

ylenediamine, N-cyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfonamido)-
 diphenylamine, N, N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-
 allyldiphenylamine, 4-isopropoxy-diphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-
 octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, e.g.
 5 p,p'-di-tert-octyldiphenyl-amine, 4-n-butylaminophenol, 4-butyrylamino-phenol, 4-nona-
 noylamino-phenol, 4-dodecanoylamino-phenol, 4-octadecanoylamino-phenol, di-(4-metho-
 xyphenyl)-amine, 2,6-di-tert-butyl-4-dimethylamino-methyl-phenol, 2,4'-diamino-diphen-
 ylmethane, 4,4'-diamino-diphenylmethane, N,N,N',N'-tetramethyl-4,4'-diamino-diphenyl-
 methane, 1,2-di-((2-methyl-phenyl)-amino)-ethane, 1,2-di-(phenylamino)propane, (o-
 10 tolyl)biguanide, di(4-(1',3'-dimethyl-butyl)-phenyl)amine, tert-octylated N-phenyl-1-naph-
 thylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mix-
 ture of mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated
 dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphen-
 ylamines, mixtures of mono- and dialkylated tert-butyldiphenylamines, 2,3-dihydro-3,3-
 15 dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- and dialkylated tert-
 butyl/tert-octyl-phenothiazines, a mixture of mono- and dialkylated tert-octyl-phenothia-
 zines, N-allylphenothiazine, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis-
 (2,2,6,6-tetramethylpiperidin-4-yl)hexamethylenediamine, bis-(2,2,6,6-tetramethyl-
 piperidin-4-yl) sebacate, 2,2,6,6-tetramethylpiperidin-4-one or 2,2,6,6-tetramethyl-
 20 piperidin-4-ol; and

17) Aliphatic or aromatic phosphites, esters of thiodipropionic acid or of thiodiacetic acid, or
 salts of dithiocarbamic or dithiophosphoric acid, 2,2,12,12-tetramethyl-5,9-dihydroxy-
 3,7,1-trithiatridecane or 2,2,15,15-tetramethyl-5,12-dihydroxy-3,7,10,14-tetrathiahexa-
 decane.

25 Examples of metal passivators, for example for copper, are:

- 1) Benzotriazoles and their derivatives, for example 4- or 5-alkylbenzotriazoles (e.g.
 tolutriazole) and derivatives thereof, 4,5,6,7-tetrahydrobenzotriazole, 5,5'-methylene-
 bisbenzotriazole; Mannich bases of benzotriazole or tolutriazole, such as 1-(di(2-
 ethylhexyl)aminomethyl)tolutriazole and 1-(di(2-ethylhexyl)aminomethyl)-benzotria-
 30 zole; alkoxyalkylbenzotriazoles, such as 1-(nonyloxymethyl)-benzotriazole, 1-(1-bu-
 toxyethyl)-benzotriazole and 1-(1-cyclohexyloxybutyl)-tolutriazole;
- 2) 1,2,4-Triazoles and derivatives thereof, for example 3-alkyl(or aryl)-1,2,4-triazoles,
 Mannich bases of 1,2,4-triazoles such as 1-(di(2-ethylhexyl)aminomethyl)-1,2,4-tria-
 zole; alkoxyalkyl-1,2,4-triazoles such as 1-(1-butoxyethyl)-1,2,4-triazole; acylated 3-
 35 amino-1,2,4-triazoles;

- 3) Imidazole derivatives, for example 4,4'-methylenebis(2-undecyl-5-methyl-imidazole), bis((N-methyl)imidazol-2-yl)carbinol octyl ether;
- 4) Sulfur-containing heterocyclic compounds, for example 2-mercaptobenzothiazole, 2,5-dimercapto-1,3,4-thiadiazole, 2,5-dimercaptobenzothiadiazole and derivatives thereof; 3,5-bis(di(2-ethylhexyl)aminomethyl)-1,3,4-thiadiazolin-2-one; and
- 5) Amino compounds, for example salicylidenepropylenediamine, salicylaminoguanidine and salts thereof.

Examples of rust inhibitors are:

- 1) Organic acids, their esters, metal salts, amine salts and anhydrides, for example alkyl- and alkenylsuccinic acids and the partial esters thereof with alcohols, diols or hydroxycarboxylic acids, partial amides of alkyl- and alkenylsuccinic acids, 4-nonylphenoxyacetic acid, alkoxy- and alkoxyethoxycarboxylic acids, such as dodecyloxyacetic acid, dodecyloxy(ethoxy)acetic acid and the amine salts thereof, and also N-oleoylsarcosine, sorbitan monooleate, lead naphthenate, alkenylsuccinic anhydrides, for example dodecenylsuccinic anhydride, 2-(2-carboxyethyl)-1-dodecyl-3-methylglycerine and its salts, especially sodium and triethanolamine salts;
- 2) Nitrogen-containing compounds, for example primary, secondary or tertiary aliphatic or cycloaliphatic amines and amine salts of organic and inorganic acids, for example oil-soluble alkylammonium carboxylates, and also 1-(N,N-bis(2-hydroxyethyl)amino)-3-(4-nonylphenoxy)propan-2-ol; or heterocyclic compounds, for example: substituted imidazolines and oxazolines, 2-heptadecenyl-1-(2-hydroxyethyl)-imidazoline;
- 3) Phosphorus-containing compounds, for example amine salts of phosphoric acid partial esters or phosphonic acid partial esters, zinc dialkyldithiophosphates;
- 4) Sulfur-containing compounds, for example: barium dinonylnaphthalene-sulfonates, calcium petroleum sulfonates, alkylthio-substituted aliphatic carboxylic acids, esters of aliphatic 2-sulfocarboxylic acids and salts thereof; and
- 5) Glycerine derivatives, for example: glycerine monooleate, 1-(alkylphenoxy)-3-(2-hydroxyethyl)glycerines, 1-(alkylphenoxy)-3-(2,3-dihydroxypropyl)glycerines, 2-carboxyalkyl-1,3-dialkylglycerines.

Examples of viscosity index improvers are:

Polyacrylates, polymethacrylates, vinylpyrrolidone/methacrylate copolymers, polyvinylpyrrolidones, polybutenes, olefin copolymers, styrene/acrylate copolymers, polyethers.

Examples of pour point depressants are:

Polymethacrylates, alkylated naphthalene derivatives.

Examples of dispersants/surfactants are:

- 5 Polybutenylsuccinamides or -imides, polybutenylphosphonic acid derivatives, and basic magnesium, calcium and barium sulfonates, phenolates and salicylates.

Examples of antifoaming agents are: silicone oils and polymethocrylen.

The demulsifiers are, for example, selected from:

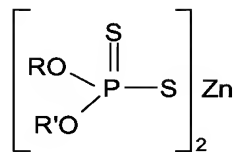
Polyetherpolyols and dinonylnaphthalenesulfonates.

The friction modifiers are, for example, selected from:

- 10 Fatty acids and their derivatives (i.e. natural esters of fatty acids such as glycerol monooleate), amides, imides and amines (i.e. oleylamine), sulfur containing organomolybdenum dithiocarbamates, sulfur-phosphorus containing organomolybdenum dithiophosphates, sulfur-nitrogen containing organomolybdenum compounds based on dispersants, molybdenum carboxylate salts, molybdenum-amine complexes, molybdenum amine/alcohol/amid complexes and molybdenum cluster compounds, Teflon® and molybdenum disulfide.
- 15

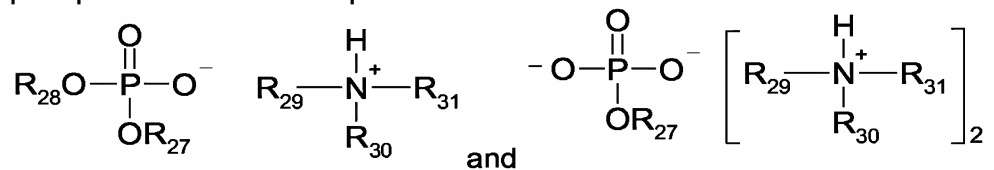
Examples of additional antiwear additives are:

- Sulfur- and/or phosphorus- and/or halogen-containing compounds, such as sulfurized olefins and vegetable oils, zinc dialkyldithiophosphates, tritolyl phosphate, tricresyl phosphate, chlorinated paraffins, alkyl and aryl di- and trisulfides, amine salts of mono- and dialkyl phosphates, amine salts of methylphosphonic acid, diethanolaminomethyltolyltriazole, di-(2-ethylhexyl)-aminomethyltolyltriazole, derivatives of 2,5-dimercapto-1,3,4-thiadiazole, ethyl(bisisopropoxyphosphinothioyl)thiopropionate, triphenyl thiophosphate (triphenyl phosphorothioate), tris(alkylphenyl) phosphorothioates and mixtures thereof (for example tris(isononylphenyl) phosphorothioate), diphenylmonononylphenyl phosphorothioate, isobutylphenyl diphenyl phosphorothioate, the dodecylamine salt of 3-hydroxy-1,3-thiaphosphetan 3-oxide, trithiophosphoric acid 5,5,5-tris-isooctyl 2-acetate, derivatives of 2-mercaptobenzothiazole, such as 1-N,N-bis(2-ethylhexyl)aminomethyl-2-mercapto-1H-1,3-benzothiazole, and ethoxycarbonyl 5-octyldithiocarbamate;
 - Dihydrocarbyl dithiophosphate metal salts where the metal is aluminum, lead, tin manganese, cobalt, nickel, zinc or copper, but most often zinc. The zinc salt (zinc dialkyl dithiophosphate) is represented as
- 20
- 25
- 30



where R and R' are independently C₁-C₂₀ alkyl, C₃-C₂₀ alkenyl, C₅-C₁₂ cycloalkyl, C₇-C₁₃ aralkyl or C₆-C₁₀ aryl, for example R and R' are independently C₁-C₁₂ alkyl;

- Antiwear additives as described in U.S. Patent Specification Nos. 4,584,021; 5,798,321; 5,750,478; 5,801,130; 4,191,666; 4,720,288; 4,025,288; 4,025,583 and in WO 095/20592, which are incorporated herein by reference; amines for example polyalkylene amines such as ethylene diamine, diethylene triamine, triethylene tetraamine, tetraethylene pentamine, pentaethylene hexamine, nonaethylene decamine and aryl amines as described in United States Patent Specification No. 4,267,063, herein incorporated by reference; salts of amine phosphates comprising specialty amines and mixed mono- and di-acid phosphates; the mono- and di-acid phosphate amines correspond to the structural formulae:



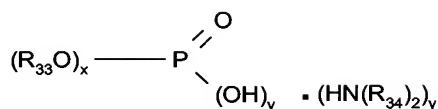
wherein R₂₇ is hydrogen, C₁-C₂₅ linear or branched chain alkyl which is unsubstituted or substituted by one or more C₁-C₆alkoxy groups, a saturated acyclic or alicyclic group, or aryl;

R₂₈ is C₁-C₂₅ linear or branched chain alkyl which is unsubstituted or substituted by one or more C₁-C₆alkoxy groups, a saturated acyclic or alicyclic group, or aryl;

R₂₉ is hydrogen, C₁-C₂₅ linear or branched chain alkyl, a saturated or unsaturated acyclic or alicyclic group, or aryl; and are hydrogen or C₁-C₁₂ linear or branched chain alkyl; and

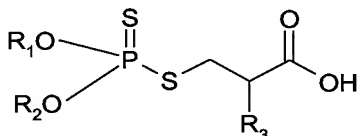
R₃₀ and R₃₁ are, each independently of the other, C₁-C₂₅ linear or branched chain alkyl, a saturated or unsaturated acyclic or alicyclic group, or aryl. Preferably, R₂₇ and R₂₈ are linear or branched C₁-C₁₂ alkyl; and R₂₉, R₃₀ and R₃₁ are linear or branched C₁-C₁₈ alkyl;

- A mixture of amine phosphates, CAS# 80939-62-4, particularly by enhancing the wear performance of the base oil such that it meets stringent military performance specifications;



wherein R_{33} is n-hexyl, R_{34} is C_{11} - C_{14} branched alkyl, and when $x=1$ then $y=2$; when $x=2$ then $y=1$;

- Other conventional antiwear additives of the formula



in which R_1 and R_2 independently of one another are C_3 - C_{18} alkyl, C_5 - C_{12} cycloalkyl, C_5 - C_6 cycloalkylmethyl, C_9 - C_{10} bicycloalkylmethyl, C_9 - C_{10} tricycloalkylmethyl, phenyl or C_7 - C_{24} alkylphenyl or together are $(\text{CH}_3)_2\text{C}(\text{CH}_2)_2$, R_3 is hydrogen or methyl.

- 5
- 10 The additives (I), (II), and (III) can be introduced into the lubricating oil in manners known per se. The compounds are readily soluble in oils. They may be added directly to the lubricating oil or they can be diluted with a substantially inert, normally liquid organic diluent such as naphtha, benzene, toluene, xylene or a normally liquid oil to form an additive concentrate or masterbatch. These concentrates generally contain from about 10% to about 90% by weight
- 15 additive and may contain one or more other additional additives. The additives may be introduced as part of an additive package.

A further embodiment of the invention relates to process for the reduction of wear in combustion engines, which comprises adding to the engine the lubricant composition as defined above. In a preferred embodiment the total amount of sulphur in that composition is less than

20 0.3%, particularly 0.2%, by weight and that of phosphorus less than 0.08% by weight.

The invention is further illustrated by the following Examples. Unless otherwise indicated, parts and percentages are by weight.

Example 1

Preparation of Ethylenediamine tetra-N-n-octylacetamide

- 25 To a stirred mixture of 2.80 g ethylenediamine, 20.06 g sodium carbonate, and 100 mg potassium iodide in 100 ml acetonitrile is added a solution of 38.35 g of 2-chloro-N-n-octylacetamide. After being stirred at ambient temperature for 18 hours, the mixture is heated at reflux for 5 days. After cooling, the mixture is partitioned between 400 ml dichloromethane and 250 ml water. The aqueous phase is further extracted with 200 ml dichloromethane. The

combined organic extract is washed with water (200 ml), brine (200 ml) and dried over anhydrous sodium sulfate. The solvent removed *in vacuo*, and the resulting residue is recrystallized from ethyl acetate to give the product as a pale yellow fibrous wax.

Example 2

5 Preparation of Ethylenediamine tetra-N,N-di-n-octylacetamide

A mixture of 78.7 g 2-chloro-N,N-dioctylacetamide, 3.7 g ethylenediamine and 26.2 g sodium carbonate in 300 ml N,N-dimethylacetamide is heated at 90°C for 3 hours and at 120°C for another 16 hours. The mixture is cooled, washed with water, extracted with hexane and dried over sodium sulfate. The hexanes are removed *in vacuo* to give 74.9 g of product as a viscous yellow liquid.

Alternative one-pot procedure:

To a rapidly stirred mixture of di-n-octylamine (25.03 g), xylene (25 ml), Na₂CO₃ (12.5 g) and water (125 ml) chloroacetyl chloride (12.4 g) is added dropwise over 20 min. Intermittent cooling is applied to maintain temperature between 20–25°C. After completion of addition, the mixture is stirred for 30 min. The phases are allowed to separate, and the lower aqueous phase is removed. N,N-Dimethylacetamide (25 ml), ethylenediamine (1.55 g), and Na₂CO₃ (12.5 g) are added, and the mixture heated with stirring to 120°C. The reaction mixture is heated for 18 hrs, allowed to cool and mixed with water (125 ml) to dissolve salts and DMAc. After removing the aqueous phase the solvents are removed *in vacuo*. The product is filtered to remove sediments, to yield 28.77 g (93%) of product as a pale, yellow oil. Xylene may be replaced with other suitable solvents, for example ethylbenzene.

Example 3

Preparation of N,N-di-n-octyl-2-(di-n-octylamino)acetamide

To a rapidly stirred solution 32.62 g di-n-octylamine in 100 ml acetonitrile, 7.62 g 2-chloroacetyl chloride is added over a 1 hour period. The mixture is stirred at ambient temperature for 3 hours, followed by the addition of 15.91 g sodium carbonate and 0.63 g potassium iodide. The mixture is heated at reflux for 24 hours. After cooling, the mixture is partitioned between 250 ml dichloromethane and 250 ml water. The aqueous phase is further extracted with 250 ml dichloromethane. The combined organic extract is washed with water (100 ml), brine (100 ml) and dried over anhydrous sodium sulfate. The solvent is removed *in vacuo* and the resulting residue is partitioned between hexane (300 ml) and acetonitrile (250 ml). After filtering to remove undissolved solids, the phases are separated, and the yel-

low hexanes phase is washed with 2 portions (100 ml each) acetonitrile. The solvent is removed *in vacuo* to give the product as a waxy orange oil.

Example 4

Preparation of Diethylenetriamine penta-N,N-di-n-octylacetamide

- 5 A mixture of 31.8 g 2-chloro-N,N-dioctylacetamide, 2.08 g diethylenetriamine and 10.6 g sodium carbonate in 150 ml N,N-dimethylacetamide is heated at 150°C for 48 hours. After cooling the mixture is washed with water, extracted with hexane and dried over sodium sulfate. The hexane is removed *in vacuo* to give 30.6 g of the product as a dark brown liquid.

Example 5

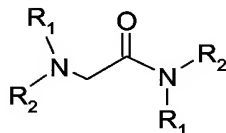
10 Preparation of Ethylenediamine tetra-N-oleylacetamide

- To a rapidly stirred mixture of oleylamine (118.5 g), diether (200 ml), Na₂CO₃ (50.29 g) and water (500 ml) 2-chloroacetyl chloride (54.16 g) is added dropwise over 60 min. Intermittent cooling is applied to maintain temperature between 10-15°C. After completion of addition, the mixture is stirred for 60 min, and the phases are allowed to separate. Analysis (H-NMR) of a sample of the upper ether layer indicates that a small amount of amine is not reacted. Additional Na₂CO₃ (7 g) and water (50 ml) is added, followed by chloroacetyl chloride (7 g). After removing the upper ether phase the aqueous phase is extracted with additional ether (200 ml). The combined organic phase is washed with water (2 x 125 ml portions), saturated NaCl (125 ml) and dried over anh. Na₂SO₄. The solvent is removed *in vacuo* to give 149 g 2-chloro-N-oleylacetamide.
- 15
- 20

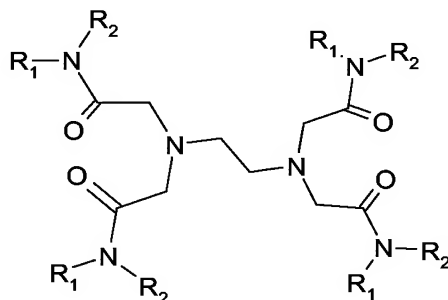
- A mixture of ethylenediamine (1.01 g), 2-chloro-N-oleylacetamide (23.13 g), N,N-dimethylacetamide (50 ml) and Na₂CO₃ (25.8 g) is heated for 20 hours at 120-130°C. After cooling, the reaction mixture is partitioned between diethyl ether (250 ml) and water (250 ml). The ether layer is washed with water (3 x 100 ml), saturated NaCl (100 ml) and dried over anh. Na₂SO₄. The solvent is removed *in vacuo* to give 20.4 g of the tetra-alkylated product.
- 25

Example 6

The following compounds are prepared according to the methods described herein.



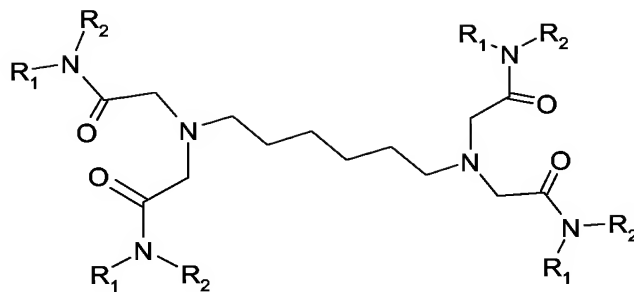
R ₁	R ₂	Physical Form
n-Octyl	n-Octyl	Liquid (Ex.3)



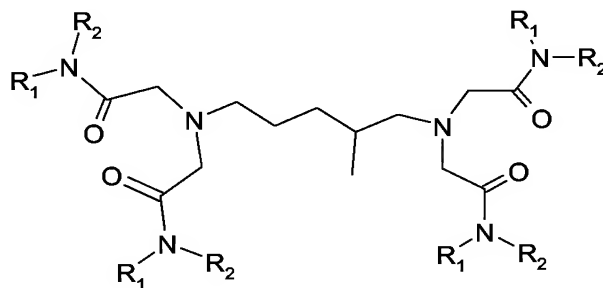
5

R ₁	R ₂	Physical Form
2-Ethylhexyl	2-Ethylhexyl	Liquid
n-Octyl	n-Octyl	Liquid (Ex. 29)
Oleyl	Hydrogen	Wax (Ex. 5)
n-Octyl	Hydrogen	Wax (Ex. 1)
t-Octyl	Hydrogen	Solid
Dodecyl	Hydrogen	Solid
C ₁₂ -C ₁₅ Alkyl	Hydrogen	Syrup
C ₁₈ -C ₂₄ Alkyl	Hydrogen	Syrup

- 20 -

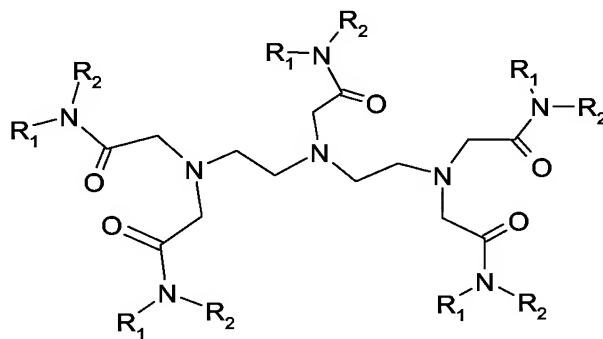


R ₁	R ₂	Physical Form
n-Octyl	n-Octyl	Liquid
t-Octyl	Hydrogen	Resin
2-Ethylhexyl	2-Ethylhexyl	Liquid



5

R ₁	R ₂	Physical Form
t-Octyl	Hydrogen	Resin



R ₁	R ₂	Physical Form
n-Octyl	n-Octyl	Liquid (Ex. 4)
2-Ethylhexyl	2-Ethylhexyl	Liquid

Example 7**Application Example**

5 Antiwear properties are measured on a PCS Instruments Mini-Traction Machine, modified with a Pin-on-Disc attachment, in which a stationary pin (500 x 500 μ) is held against a rotating disc. A fixed load is applied at a constant temperature. Wear is measured as the displacement of the pin, due to loss of material from the pin. The test oil is a zero S, very low P automotive engine oil, fully formulated except that no antiwear additive is included. The reaction test conditions are 10N load, oil temperature 100°C. Wear data is recorded for 60 min, and the average wear rate is reported here as the linear regression slope of the wear curve.

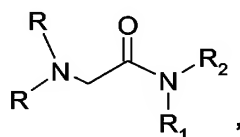
Oil	Wear Rate
Test oil (no antiwear additive)	279 μ /hr
Test oil + 1.2% ZDDP	3.0 μ /hr
Test oil + 1% additive of Example 2	16 μ /hr

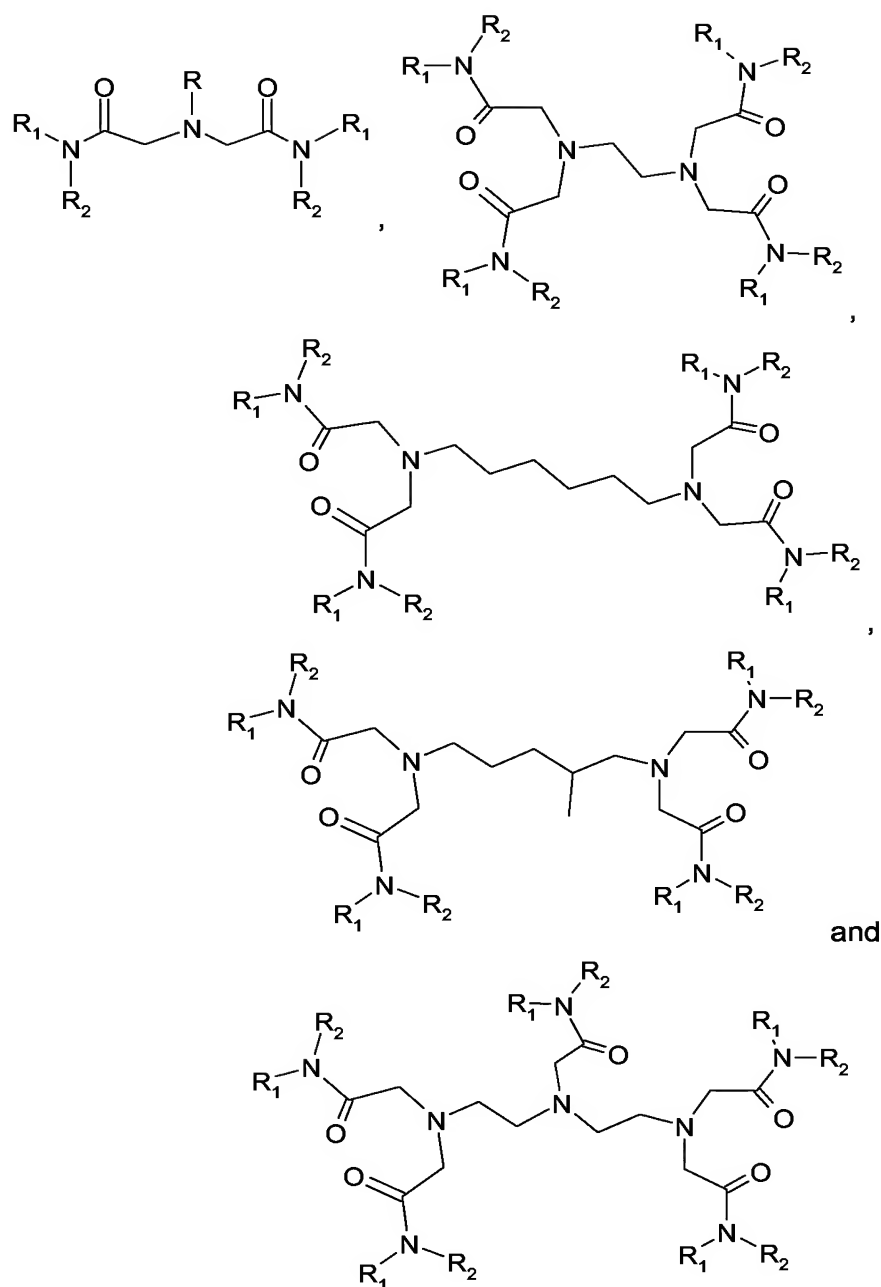
10 ZDDP is zinc dialkyl dithiophosphate. A secondary ZDDP at 1.2% provides 0.1% P.

each amide nitrogen atom is either mono- or disubstituted by alkyl or alkenyl; and that the alkyl or alkenyl groups have 8 to 24 carbon atoms, if the amide nitrogen is monosubstituted by alkyl or alkenyl; and

that the carbon atoms of the alkyl or alkenyl groups have 8 to 18 carbon atoms, if the amide nitrogen is disubstituted by alkyl or alkenyl.

- 5
2. A composition according to claim 1, which comprises at least one compound selected from the group consisting of N-alkylaminoacetamide compounds (I) and (II), wherein the total number of carbon atoms of the alkyl or alkenyl groups are from 14 to 18 carbon atoms if the amide nitrogen is disubstituted by alkyl or alkenyl.
- 10
3. A composition according to claim 1, which comprises at least one compound selected from the group consisting of alkylenedi-, alkylenetri- and alkylenetetra-amine acetamide compounds (III), wherein A and G are selected from the group consisting of ethylene, propylene, hexamethylene and 2-methylpentylene.
- 15
4. A composition according to claim 1, wherein each R' is independently straight or branched chain alkyl or alkenyl of 7 to 9 carbon atoms.
5. A composition according to claim 1, wherein one R' is hydrogen and the other one is straight or branched chain alkyl or alkenyl of 14 to 18 carbon atoms.
6. A composition according to claim 1, wherein each R' is identical and is straight or branched chain alkyl or alkenyl of 7 to 9 carbon atoms.
- 20
7. A composition according to claim 1, wherein the compounds (I), (II) and (III) are selected from the group consisting of

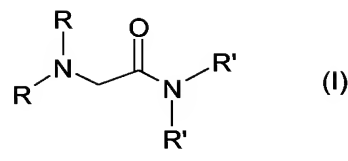




- 5 wherein R is n-octyl and each of R_1 and R_2 are 2-ethylhexyl or n-octyl or one of R_1 and R_2 is hydrogen and the other is oleyl, n-octyl, t-octyl or dodecyl.

8. An alkylaminoacetamide compound (I)

- 25 -



Wherein

R, each independently, is alkyl or alkenyl of 1 to 8 carbon atoms and

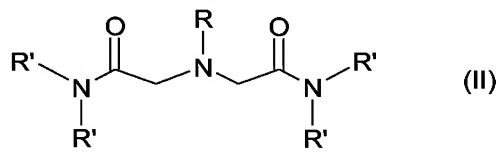
R', each independently, is hydrogen or alkyl or alkenyl or 1 to 24 carbon atoms,

5 provided that

each amide nitrogen atom is either mono- or disubstituted by alkyl or alkenyl; and that the alkyl or alkenyl groups have 8 to 24 carbon atoms, if the amide nitrogen is mono-substituted by alkyl or alkenyl; and

10 that the alkyl or alkenyl groups have 8 to 18 carbon atoms, if the amide nitrogen is disubstituted by alkyl or alkenyl.

9. An N-alkylaminoacetamide compound (II)



Wherein

R, each independently, is alkyl or alkenyl of 1 to 8 carbon atoms and

15 R', each independently, is hydrogen or alkyl or alkenyl or 1 to 24 carbon atoms,

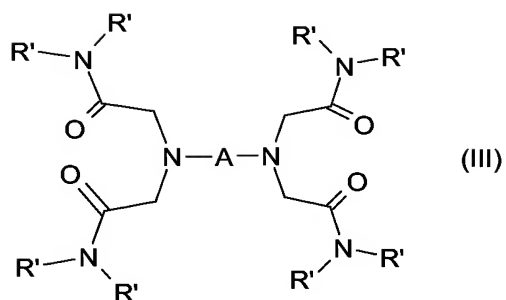
provided that

each amide nitrogen atom is either mono- or disubstituted by alkyl or alkenyl; and that the alkyl or alkenyl groups have 8 to 24 carbon atoms, if the amide nitrogen is mono-substituted by alkyl or alkenyl; and

20 that the alkyl or alkenyl groups have 8 to 18 carbon atoms, if the amide nitrogen is disubstituted by alkyl or alkenyl.

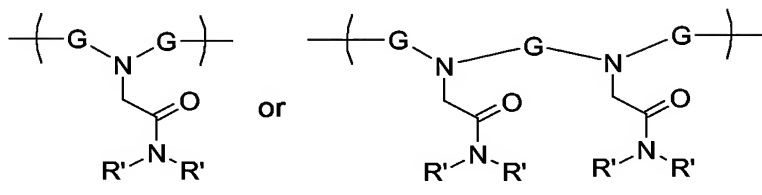
10. An alkylenedi-, alkylenetri- or alkylenetetra-amine acetamide compound (III)

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wherein

A is alkylene of from 2 to 6 carbon atoms or is a group



5 G, each independently, is alkylene of 2 to 6 carbon atoms,

R', each independently, is alkyl or alkenyl of 1 to 24 carbon atoms, where for each amide group, the total number of carbon atoms of the alkyls or alkenyl groups are from 14 to 18 carbon atoms.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/054347

A. CLASSIFICATION OF SUBJECT MATTER

C10M133/16 C07C237/06

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C10M C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, BEILSTEIN Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2 191 978 A (BALLE GERHARD ET AL) 27 February 1940 (1940-02-27) column 1, line 1 - line 40 claims; examples 4,5	8
X	US 2 516 674 A (BRUCE WILLIAM F ET AL) 25 July 1950 (1950-07-25) column 1, line 4 - line 11 claims; examples	8
X	US 2 548 863 A (BRUCE WILLIAM F ET AL) 17 April 1951 (1951-04-17) column 1, line 24 - line 55 claims	8
	----- -/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search

18 November 2005

Date of mailing of the international search report

25/11/2005

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/054347

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4 358 387 A (ZOLESKI ET AL) 9 November 1982 (1982-11-09) column 1, line 40 - line 43 column 2, line 60 - column 3, line 14 claims; examples -----	1-10
A	US 2 411 662 A (MARTIN HENRY ET AL) 26 November 1946 (1946-11-26) the whole document -----	1-10

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP2005/054347

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 2191978	A	27-02-1940	NONE	
US 2516674	A	25-07-1950	NONE	
US 2548863	A	17-04-1951	NONE	
US 4358387	A	09-11-1982	NONE	
US 2411662	A	26-11-1946	NONE	